

# Measurement of scintillation and ionization yield with high-pressure gaseous mixtures of Xe and TMA for improved neutrinoless double beta decay and dark matter searches

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**ABSTRACT:** The gaseous Xenon(Xe) time projection chamber (TPC) is an attractive detector technique for neutrinoless double beta decay and WIMP dark matter searches. While it is less dense compared to Liquid Xe detectors, it has intrinsic advantages in tracking capability and better energy resolution. The performance of gaseous Xe can be further improved by molecular additives such as trimethylamine(TMA), which is expected to (1) cool down the ionization electrons, (2) convert Xe excitation energy to TMA ionizations through Penning transfer, and (3) produce scintillation and electroluminescence light in a more easily detectable wavelength (300 nm). In order to test the feasibility of the performance improvements with TMA, we made the first direct measurement of Penning and fluorescence transfer efficiency with gaseous mixtures of Xe and TMA. While we observed a Penning transfer efficiency up to  $\sim 35\%$ , we found strong suppression of primary scintillation light with TMA. We also found that the primary scintillation light with Xe and TMA mixture can be well characterized by  $\sim 3\%$  fluorescence transfer from Xe to TMA, with further suppression due to TMA self-quenching. No evidence of the scintillation light produced by recombination of TMA ions was found. This strong suppression of scintillation light makes dark matter searches quite challenging, while the possibility of improved neutrinoless double beta decay searches remains open. This work has been carried out within the context of the NEXT collaboration.

**KEYWORDS:** Gaseous TPC; Xe; TMA; Neutrinoless double beta decay; Dark matter; NEXT.

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## 1. Introduction

Searches for WIMP dark matter and neutrinoless double beta decay are currently among the most important tasks in nuclear and particle physics. Time projection chambers (TPCs) with Xenon (Xe) as a target and detection medium are commonly used for the searches with leading sensitivities. While liquid Xe is more commonly used for such applications, gaseous Xe has intrinsic advantages over liquid Xe. The NEXT experiment [1] is searching for neutrinoless double beta decay with the superior energy resolution and tracking capabilities of gaseous Xe TPCs. In addition, there is a potential capability of performing dark matter searches with directional sensitivity utilizing columnar recombination [2, 3].

We are exploring possible performance improvements for gaseous Xe TPCs by adding trimethylamine(TMA). We report measurements of ionization and scintillation yields with a new ionization chamber that we have recently commissioned at Lawrence Berkeley National Laboratory.

## 2. Performance improvements with TMA

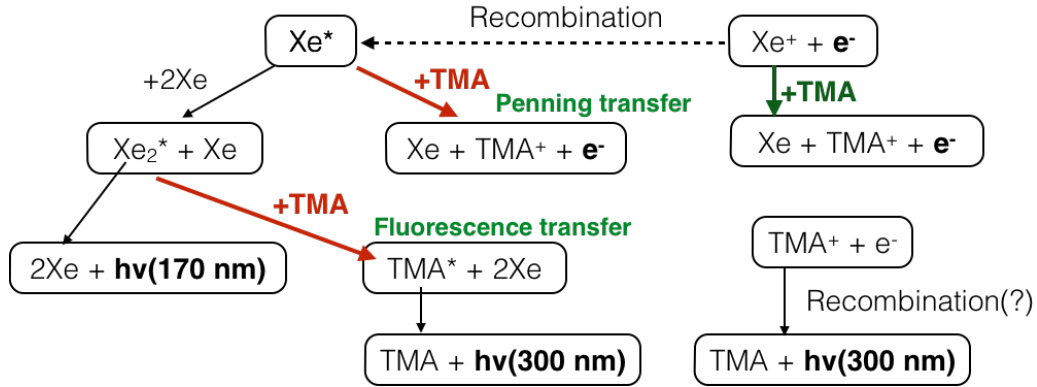
There are two major categories of performance improvements with TMA. The first one is the efficient cooling of ionization electrons due to its large inelastic cross section. This significantly reduces the amount of diffusion and improves tracking performances for neutrinoless double beta decay searches [4]. The reduction of electron diffusion also enhances charge recombination processes between any ions and electrons, which is essential for measuring the direction of short tracks using columnar recombination [3].

The other category of improvements is a transformation of observable signal through Xe and TMA interactions. Figure 1 shows a schematics of Xe and TMA interactions after initial excitation and ionization of Xe. For pure Xe detectors, typical observable signals are scintillation light from

excited Xe ( $\text{Xe} \rightarrow \text{Xe}^*$ ,  $\text{Xe}^* + 2\text{Xe} \rightarrow \text{Xe}_2^* + \text{Xe}$ ,  $\text{Xe}_2^* \rightarrow 2\text{Xe}(g.s.) + h\nu(170 \text{ nm})$ ) and ionization electrons ( $\text{Xe} \rightarrow \text{Xe}^+ + e^-$ ). With the addition of TMA, we expect to observe (1) Penning transfer that convert excitations of Xe into ionizations of TMA ( $\text{Xe}^* + \text{TMA} \rightarrow \text{Xe} + \text{TMA}^+ + e^-$ ), and (2) the fluorescence transfer that convert excitations of Xe to excitations of TMA, which then de-excite by emitting  $\sim 300 \text{ nm}$  light, which is much more easily detectable with PMTs compared to the light from Xe at  $\sim 170 \text{ nm}$ . Those excited and ionized TMA molecules are also produced by direct interactions between incoming particles and TMA. Their contribution to the total observable signal would be marginal at a typical TMA concentration of  $\sim 1\%$ , however. It is also important to note that most the observable light would come from TMA de-excitation, because TMA absorbs  $170 \text{ nm}$  light very efficiently, while it is practically transparent to its own light at  $\sim 300 \text{ nm}$  [4].

Those processes, together with the charge exchange process ( $\text{Xe}^+ + \text{TMA} \rightarrow \text{Xe} + \text{TMA}^+$ ), would convert initial  $\text{Xe}^*$  and  $\text{Xe}^+$  into  $\text{TMA}^*$  and  $\text{TMA}^+$  with an increased number of ions. The energy resolution is impacted by the two competing mechanisms; (1) a decrease due to the averaging effect of the Penning transfer reactions [5], and (2) an increase due to the fluctuations in the former process as well as the additional fluctuations in the direct ionization, introduced through the new inelastic degrees of freedom of the molecular additive [6]. They both compose a new Fano factor for the mixture. The average number of ionizations  $n_o$  (that also contributes to the resulting energy resolution as  $\sqrt{F/n_o}$ ) can be generally expected to increase for Penning mixtures, except for very high concentrations of the additive, and can be accessed directly through current measurements, as those reported here. If recombinations of ionized TMA produce similar fluorescence light via  $\text{TMA}^+ + e^- \rightarrow \text{TMA}^* \rightarrow \text{TMA}(g.s.) + h\nu(300 \text{ nm})$ , we can measure the size of columnar recombination and obtain directional sensitivity.

However, the efficiency of each processes with Xe+TMA mixture was poorly known. In order to realistically estimate the improvement in performance, we made the first direct measurement of the Penning transfer and the fluorescence transfer efficiencies.

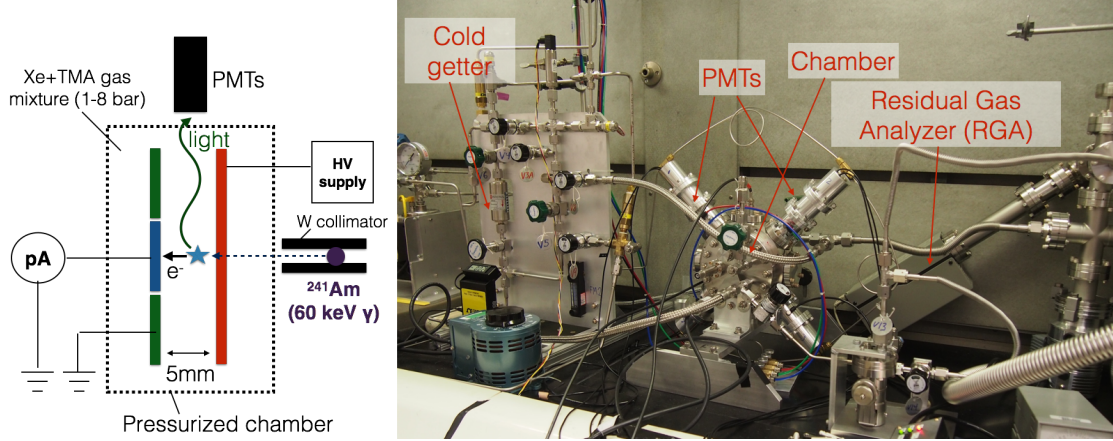


**Figure 1.** Simplified schematic of Xe and TMA reactions after initial ionization and excitation of Xe. We made the first direct measurement of the processes shown with red arrows.

### 3. Experimental setup

A schematic of the ionization chamber and its photograph are shown in figure 2. A pair of parallel

plate electrodes with gap width of 5 mm is housed in a pressurized gas chamber. The charge is collected in the inner electrode with 2.5 cm diameter. Four Hamamatsu R7378A PMTs, which have sensitivity to VUV photons down to  $\sim 150$  nm, are placed at the periphery of the electrodes and used to detect scintillation light from the gas between the electrodes. The gas between the electrodes was irradiated with  $\sim 60$  keV gamma rays from a 10 mCi  $^{241}\text{Am}$  radioactive source. Currents from electrodes and PMTs were read with pico-ammeters in DC mode. All the measurements were done in a room at the temperature of  $(20 \pm 2)^\circ\text{C}$ . A more detailed description of the setup can be found in Ref. [7].



**Figure 2.** Schematic of the ionization chamber (left) and a photograph of the setup (right).

## 4. Results

After extensively testing the system with pure Xe gas and confirming the detector performance [7], we introduced a gaseous mixture of Xe and TMA into the system. We describe the results of primary charge and primary scintillation yield in this section, improved from our initial results [8].

### 4.1 Primary charge yield

Figure 3 shows the charge yield observed at the inner electrode at approximately 4 bar total pressure and for various TMA concentrations given in the volume fraction. At the medium electric field region of  $10^3 < E/\rho < 10^5$  ( $\text{Vcm}^2\text{g}^{-1}$ ), we found a slightly larger electric field dependence of the charge yield at higher concentration of TMA. That is interpreted as an effect of increased recombination, which reduces the charge yield at a higher TMA concentration and at a lower electric field. At the high electric field of  $\sim 10^5$   $\text{Vcm}^2\text{g}^{-1}$ , where the recombination effect is negligible, we observed an increase of the charge yield as expected by the Penning transfer. We also observed charge amplification starting at  $E/\rho > 2 \times 10^5$   $\text{Vcm}^2\text{g}^{-1}$  for the Xe+TMA mixtures, whose threshold is lower than the case of pure Xe because of the lower ionization potential of TMA.

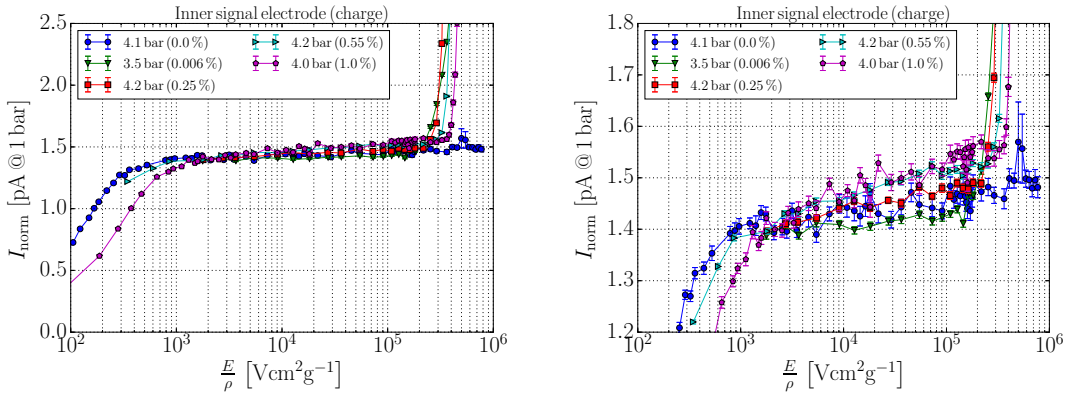
The left panel of figure 4 shows the averaged charge yield for  $10^5 < E/\rho < 2 \times 10^5$  ( $\text{Vcm}^2\text{g}^{-1}$ ) as a function of TMA concentration, taken at various total pressures. We found that the charge

yield is increased by 5-15% with  $\sim 1\%$  TMA concentration. Penning efficiency,  $\varepsilon(\text{Penning})$ , can be extracted as,

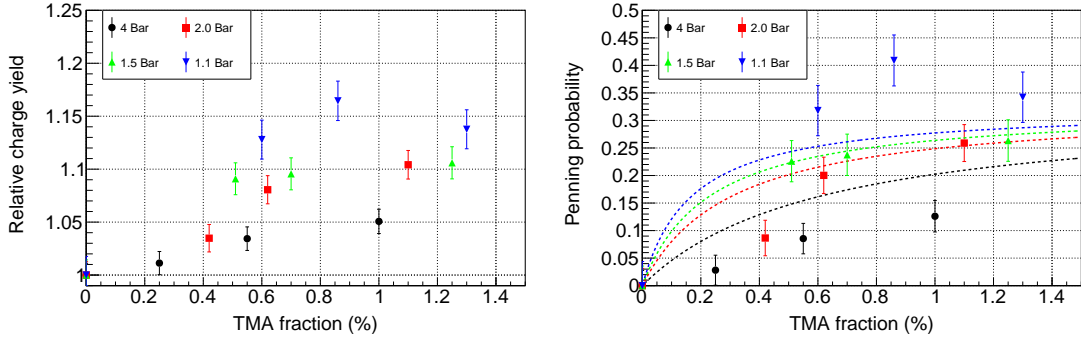
$$\varepsilon(\text{Penning}) = \frac{W_{sc}}{W_i} \left( \frac{I(\text{Xe+TMA})}{I(\text{Xe})} - 1 \right), \quad (4.1)$$

where  $W_{sc(i)}$  is electron energy loss per single excited (ionized) Xe atom, and  $I(\text{Xe})$  and  $I(\text{Xe+TMA})$  are the observed charge yield with pure Xe and Xe+TMA mixtures.

Based on the results reported in Ref. [9], the ratio  $W_{sc}/W_i$  is estimated to be  $2.5 \pm 0.8$ . The right panel of figure 4 shows the extracted value of the Penning transfer efficiencies. The error bars in the figure include the statistical uncertainties and the systematic uncertainties due the variation of the ground level and the room temperature, but do not include the global  $\sim 30\%$  uncertainty associated to  $W_{sc}/W_i$ . The results are roughly consistent with a previous indirect measurement using charge amplification in a Micromegas [5].



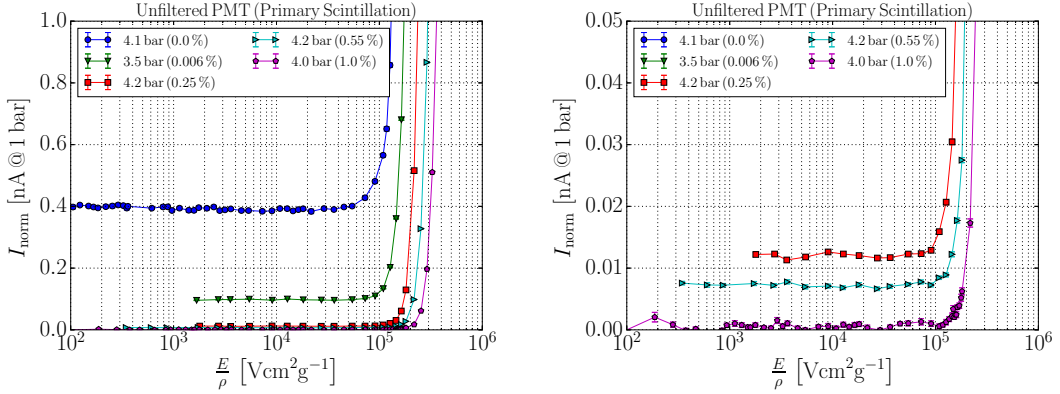
**Figure 3.** Charge yield of gas mixture of TMA and Xe as a function of external electric field for a total pressure of approximately 4 bar and various TMA concentrations. The figure on the right is the left figure with an expanded vertical scale.



**Figure 4.** Averaged primary charge yield at  $10^5 < E/\rho < 2 \times 10^5$  ( $\text{Vcm}^2\text{g}^{-1}$ ) for various Xe and TMA mixtures. The left panel shows the relative charge yield normalized to the yield with pure Xe, and the right panel shows the extracted Penning transfer probability assuming  $W_{sc}/W_i = 2.5$  [9]. The error bars show both systematic and statistical uncertainties, except for the global  $\sim 30\%$  uncertainty for the  $W_{sc}/W_i$  ratio. The dashed lines show the prediction based on the indirect measurement using a Micromegas [5].

## 4.2 Primary scintillation yield

Figure 5 shows results of light yield measurements at approximately 4 bar total pressure and for various TMA concentrations. We found the primary scintillation light is significantly reduced once a small fraction of TMA, as low as  $\sim 0.01\%$ , is introduced. We also found no evidence of the scintillation light associated with the recombination processes, which, if present, would increase the light yield at lower electric field. This suggests that, the excess energy from the recombinations, even if they occur, is predominantly released by some mechanism other than fluorescence light emissions. The increase of the light yield at  $E/\rho > \times 10^5 \text{ Vcm}^2\text{g}^{-1}$  is due to amplification processes by energized electrons. Similar amounts of light from Xe+TMA mixtures were also observed by a PMT with a long-pass filter which blocks light shorter than 250 nm. This is consistent with our expectation that the observed light from the Xe+TMA mixtures is primarily the  $\lambda \sim 300 \text{ nm}$  light from the TMA de-excitation, as described in the previous section.



**Figure 5.** Primary scintillation light yield with Xe+TMA gas mixture, measured at approximately 4 bar total pressure and various TMA concentration. The figure on the right is the left figure with an expanded vertical scale.

Figure 6 shows results of the primary scintillation measurements. We evaluate the relative light yield,

$$Y_{sc} = \frac{S_1(\text{Xe+TMA})/\epsilon_{det}(300\text{nm})}{S_1(\text{Xe})/\epsilon_{det}(170\text{nm})}, \quad (4.2)$$

where  $S_1(\text{Xe})$  and  $S_1(\text{Xe+TMA})$  are the observed amount of light for pure Xe and Xe+TMA mixtures, and  $\epsilon_{det}(\lambda)$  is the detection efficiency for the light at wavelength of  $\lambda$ . The ratio of detection efficiencies between 300 nm and 170 nm is estimated to be  $\epsilon_{det}(300\text{nm})/\epsilon_{det}(170\text{nm}) = 2.0 \pm 0.8$ , based on expected wavelength dependence of the PMT quantum efficiency and the transmittance of the view ports which separate the chamber and the PMTs.  $Y_{sc}$  can be simply modeled as a function of TMA concentrations ( $c$ ) and total pressures ( $P_{tot}$ ) [4, 10] as,

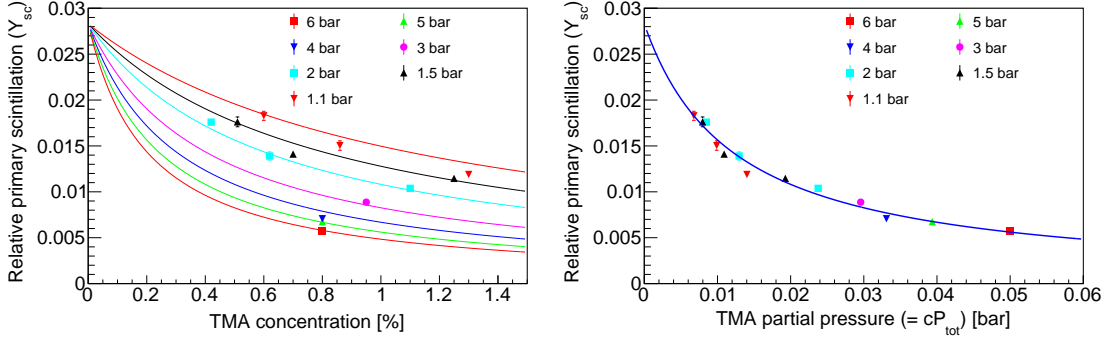
$$Y_{sc}(c, P_{tot}) = [(1 - c)P_F + c \cdot R_{sc}] \cdot \frac{1/\tau_0}{1/\tau_0 + k_{SQ}cP_{tot}}, \quad (4.3)$$

where  $P_F$  is fluorescence transfer probability,  $R_{sc} = W_{sc, Xe}/W_{sc, TMA}$  is relative amount of direct excitation of TMA,  $\tau_0 = 44 \text{ nsec}$  [10] is the decay time of the TMA excited state at the zero-



pressure limit. The  $k_{SQ}$  is the self-quenching constant that characterizes the strength of quenching the excitation of TMA by  $\text{TMA}^* + \text{TMA}$  interactions.

By simultaneously fitting the observed primary scintillation yields at different TMA concentration and pressures using Eq. (4.3), we obtained the best fit values of  $P_F = 0.03 \pm 0.01$ ,  $R_{sc} < 0.1$  and  $k_{SQ} \simeq (1.8 \pm 0.1) \times 10^9 [\text{bar}^{-1} \text{sec}^{-1}]$ . The uncertainty includes both statistical and systematic uncertainties, and is dominated by the systematic uncertainties due to temperature and the PMT dark-current variations and the uncertainty of  $\epsilon_{det}(300\text{nm})/\epsilon_{det}(170\text{nm})$ . The uncertainty for  $\epsilon_{det}(300\text{nm})/\epsilon_{det}(170\text{nm})$  is common among all the data points, while other uncertainties are assumed to be completely uncorrelated. We found that the observed primary scintillation can be well described with the fluorescence transfer from Xe to TMA at relatively low efficiency of  $\sim 3\%$  with the further reduction due to the self-quenching of the TMA. The observed value of the self-quenching constant,  $k_{SQ}$ , is within 50% of the value reported in Ref. [10]. As demonstrated in the right panel of figure 6, in the limit of low TMA concentration ( $c \ll 1$ ) and no contribution from the direct excitation of TMA ( $R_{sc} = 0$ ),  $Y_{sc}$  can be approximated as a simple function of TMA partial pressure ( $= cP_{tot}$ ), as  $Y_{sc} \simeq P_F/\tau_0/(1/\tau_0 + k_{SQ}cP_{tot})$ .



**Figure 6.** Primary scintillation yield for various Xe and TMA mixtures, as a function of TMA concentration (left) and as a function of TMA partial pressure (right). The points show the observed data, and the solid curves show the result of simultaneous fitting for all the data points with the model described in the text. The error bars show both systematic and statistical uncertainties, except for the common  $\sim 40\%$  uncertainty for the  $\epsilon_{det}(300\text{nm})/\epsilon_{det}(170\text{nm})$  ratio.

## 5. Summary

We successfully made the first direct measurement of the Penning transfer and the fluorescence transfer efficiencies of a gaseous mixture of Xe and TMA. We observed an enhancement of the charge yield up to  $\sim 15\%$  due to the Penning effect, that corresponds to a Penning transfer efficiency of up to  $\sim 35\%$ . A significant reduction of primary scintillation light with an addition of TMA was also observed. We found that the observed scintillation light can be well described assuming a fluorescence transfer from Xe to TMA at  $\sim 3\%$  efficiency and the self-quenching of TMA. We found no significant evidence of the scintillation light from TMA charge recombination.

For detection of WIMP-recoiled nucleus at  $O(10)$  keV, it is critical to precisely measure the amount of primary scintillation light in order to discriminate signals from backgrounds. This large

reduction of the primary scintillation light with TMA would make its photon statistics extremely low, and therefore makes WIMP dark matter searches challenging.

On the other hand, it is less critical for searches for neutrinoless double beta decay at  $Q \simeq 2.5$  MeV, because the initial amount of primary scintillation would be larger and also it is mainly used just for event timing measurements. Such measurements can still benefit from improved tracking performances due to smaller electron diffusion.

We demonstrated that our ionization chamber works very well for measuring scintillation and ionization yields for various gas mixtures. We are preparing a publication for our chamber system and the final results of the measurements with Xe and TMA.

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